

Citation for published version:

Hofman-Caris, R & Hofman, J 2018, Limitations of conventional drinking water technologies in pollutant removal. in A Gil, LA Galeano & MA Vincente (eds), *Applications of Advanced Oxidation Processes (AOPs) in Drinking Water Treatment*. The Handbook of Environmental Chemistry, vol. 67, Springer Verlag, Cham, Switzerland, pp. 21-51. https://doi.org/10.1007/698_2017_83

DOI:

[10.1007/698_2017_83](https://doi.org/10.1007/698_2017_83)

Publication date:

2018

Document Version

Peer reviewed version

[Link to publication](#)

This is a post-peer-review, pre-copyedit version of an chapter published in Hofman-Caris, R & Hofman, J 2018, Limitations of conventional drinking water technologies in pollutant removal. in A Gil, LA Galeano & MA Vincente (eds), *Applications of Advanced Oxidation Processes (AOPs) in Drinking Water Treatment*. The Handbook of Environmental Chemistry, vol. 67, Springer Verlag, Cham, Switzerland, pp. 21-51. The final authenticated version is available online at: https://link.springer.com/chapter/10.1007%2F698_2017_83

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Limitations of conventional drinking water technologies in pollutants removal

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Abstract

This chapter tries to give an overview of the more traditional drinking water treatment from ground and surface waters. Water is treated to meet the objectives of drinking water quality and standards. Water treatment and water quality are therefore closely connected.

The objectives for water treatment are to prevent acute diseases by exposure to pathogens, to prevent long-term adverse health effects by exposure to chemicals and micropollutants, and finally

to create a drinking water that is palatable and is conditioned in such a way that transport from the treatment works to the customer will not lead to quality deterioration.

Traditional treatment technologies as described in this chapter are mainly designed to remove macro parameters such as suspended solids, natural organic matter, dissolved iron and manganese etc. The technologies have however only limited performance for removal of micropollutants. Advancing analytical technologies and increased and changing use of compounds however show strong evidence of new and emerging threats to drinking water quality. Therefore, more advanced treatment technologies are required.

Abbreviations

AC	Activated Carbon
AOC	Assimilable Organic Carbon
BB	Building Block
BP	Biopolymer
°D	German Degree
DAF	Dissolved Air Flotation
DOC	Dissolved Organic Carbon
E2	17-beta-estradiol
EC	European Commission
<i>E. Coli</i>	<i>Escherichia Coli</i>
EDC	Endocrine Disrupting Compound
EE2	17-alpha-ethinylestradiol
FEEM	Fluorescence Excitation Emission Matrix
GAC	Granular Activated Carbon
HS	Humic Substance
LC-OCD	Liquid Chromatography – Organic Carbon Detection
LMw	Low Molecular Weight
LP	Low Pressure (UV lamp, 253.7 nm)
LRV	Logarithmic Reduction Values
LSI	Langelier Saturation Index
MP	Medium Pressure (UV lamp, 200-300 nm)
Mw	Molecular weight
NOM	Natural Organic Matter
PAC	Powdered Activated Carbon
PACl	Poly Aluminum Chloride
PCCPP	Practical Calcium Carbonate Precipitation Potential
PRAM	Polarity Rapid Assessment Method
QMRA	Quantitative Microbial Risk Assessment
REACH	Registration, Evaluation and Authorization Chemicals
SAX	Strong Anion Exchanger
SDWA	Safe Drinking Water Act
SEC	Size Exclusion Chromatography
SI	Saturation Index
SMP	Soluble Microbiological Product
SPE	Solid Phase Extraction
SUVA	Specific UV Absorbance
TCCPP	Theoretical Calcium Carbonate Precipitation Potential
TH	Total Hardness
TOC	Total Organic Carbon
TTC	Threshold of Toxicological Concern

UV	Ultra Violet
UV-A	315 – 380 nm
UV-B	280 - 315 nm
UV-C	200 – 280 nm
VUV	Vacuum Ultra Violet (100-200 nm)
WHO	World Health Organization
WWTP	Wastewater Treatment Plant

1 Safe drinking water quality

1.1 General aspects

The availability of safe drinking water is of utmost importance for human health, everywhere in the world. For centuries people have been aware of this, although they didn't always realize what caused the problems, and why the methods they applied were effective. Sanskrit texts dating from about 2000 BC indicate that drinking water should be irradiated by sunlight and afterwards filtrated over charcoal. In case the source water was unclean, it should first be boiled, then a piece of copper would have to be immersed for seven times, and finally the water would have to be filtrated. It took until about 1700 AD until Antony van Leeuwenhoek discovered microorganisms, although by that time no one yet understood the importance of this finding for human health. In 1854 during a cholera epidemic in London, John Snow, an English physician, realized that the source of the epidemic was a public water pump on Broad Street. By disabling the pump the outbreak was stopped. Since this time, the importance of safe drinking water has become more and more clear, and since the end of the 19th and beginning of the 20th century distribution of safe drinking water has become an important task of authorities. By the same time measurements were taken to improve sewerage. The combination of these two facts resulted in a significant increase in health and average lifespan of people.

As becomes clear from the above, the microbiological safety of drinking water is a very important parameter in drinking water treatment. However, it also became clear that the chemical composition of the water may play an important role. Some Roman emperors are notorious because of their mental health. This probably was caused by the fact that they were rich enough to afford drinking cups containing lead, as a result of which they obtained a very high dose of lead. Originally, drinking water mains also contained high lead concentrations, but as became clear this may cause problems for public health the mains have been replaced by polyvinyl chloride or polyethelene materials. Nowadays in some areas, like Bangladesh, high arsenic concentrations cause serious health problems. In 1989 in the Netherlands bentazone, a pesticide, was detected in drinking water. As during the last quarter of the 20th century analytical techniques were significantly improved it became clear that sources for drinking water often contained (too) high concentrations of pesticides. And shortly after it also was observed that many other organic micropollutants, like pharmaceuticals, personal care products, flame retardants, solvents and many other industrial chemicals, may be present in sources for drinking water.

Nowadays, the World Health Organization (WHO) forms an authoritative basis for the setting of national regulations and standards for water safety in support of public health. Because of the "precautionary principle", in many cases these regulations and standards are set lower than the WHO guidelines. The objective of the EU Drinking water directive (98/83/EC) is to protect human health from adverse effects of any contamination of water intended for human consumption by ensuring that it is wholesome and clean. Member States of the European Union can include additional requirements e.g. regulate additional substances that are relevant within their territory or set more stringent standards. In the USA drinking water has to comply with the Safe Drinking Water

Act (SDWA). Here standards are set and in some cases treatment technology is prescribed. The number of compounds in water seems to increase, partly because of the improvement of analytical techniques, increasing the number of detectable compounds and partly because the number of chemicals used in practice is increasing. Although there already are numerous standards set, it is likely that number of standards will further increase. Water treatment techniques aim at safe drinking water, which contains no pathogens or toxic substances. Besides, the water has to be palatable, clear, colourless and odourless. Furthermore, it should not be corrosive (for tanks and pipes), and have a low organic content, in order to prevent biological growth in pipes and tanks. Nowadays, for customer convenience and environmental reasons (decreased use of detergents and energy requirements), water often is softened and conditioned. Finally, all these requirements should be met at low costs, as drinking water is a first necessity for life. In this way it is tried to ensure safe drinking water and to keep customers trust.

1.2 Microbiological standards

Microbiological standards are important because when drinking water contains pathogens, exposure to these microorganisms can lead to acute infectious diseases and illness and in severe cases even to death. Symptoms are gastroenteritis, diarrhoea, inflammations etc. In more severe conditions, water borne diseases like typhoid fever, cholera, or poliomyelitis can occur.

Roughly three categories of pathogens with a high health impact can be distinguished: bacteria viruses and protozoa. Sometimes a fourth category is added: Helminths. Bacteria have in general a low resistance to chlorine or other disinfectants, and persistence in water supply is moderate. Some bacteria may multiply in drinking water and their infectivity is in general low to moderate. Viruses are moderately resistant to chlorine and have a high infectivity. Protozoa are highly infective and very resistant to chlorine [1].

For microbiological standards two approaches are used. The more traditional approach is the use of faecal indicators in drinking water. The standards in the European Drinking Water Directive are 0/100 mL for *E.Coli* and Enterococci [2]. However, for the more infective viruses and protozoa this approach is insufficient and a health based Quantitative Microbial Risk Assessment (QMRA) is required to determine the microbiological safety of drinking water. The QMRA methodology requires information on exposure, expressed as the number of microorganisms ingested, and dose-response models to determine the infection probability. Finally, epidemiological data can be used to determine the disease effects and severity. Using these data, the QMRA can be used to determine the health effects of drinking water at very low concentrations of pathogens. For viruses the acceptable concentrations are extremely low (1 microorganism in a few hundred to thousands m³) and thus are not measurable. Instead, logarithmic reduction values (LRV) for pathogens by individual treatment steps are used. These LRVs can be added for all individual steps in a treatment. If the number of pathogens in the raw water is known, it can be calculated by the LRV what the concentration in the treated water will be and whether this imposes an acceptable health risk.

1.3 Chemical water quality

Chemical water quality relates to several organic and inorganic compounds, both from natural and from anthropogenic origin, that may be present in drinking water. Problems caused by the presence of heavy metals have been recognized for quite some time now, and measurements have been taken to prevent these problems. Drinking water mains are no longer made of lead, and the water is conditioned (i.e. the pH is adjusted) in order to prevent dissolution of e.g. copper or carbonate from cement pipes. However, it is well known that in some parts of the world sources for drinking water contain too high concentrations of e.g. arsenic or chromium, which still causes a lot of human health

problems worldwide. Coagulation/flocculation and adsorption, sometimes in combination with oxidation can be applied to remove these metals from drinking water, and new techniques are being developed [3, 4]. For arsenic the WHO standard is 10 µg/L, but it is known that this isn't a "safe" concentration, as arsenic is a very toxic metal. However, in some cases it is technically difficult or too expensive to further decrease the arsenic concentration.

Pesticides (**Error! Reference source not found.**) are applied in both municipal and agricultural areas, and end up in sources for drinking water either by run off to surface water or by penetration into groundwater. They are organic compounds which have been designed to be harmful for certain organisms. Therefore, in principle they shouldn't be present in drinking water. However, as analytical techniques are optimized to be able to detect lower and lower concentrations, it is very difficult to ensure that "no" pesticides are present. Because of this often the "threshold of toxicological concern" (TTC) is applied, which gives a guideline for safe concentrations for certain compounds, below which no negative effects on human health are expected. In The Netherlands a standard has been set at 0.1 µg/L for individual pesticides, with a total concentration < 0.5 µg/L.



Fig. 1 Several pesticides

In the past decade it has become known that sources for drinking water also may contain pharmaceuticals. These too are compounds designed for their effect on living organisms. The major part of these pharmaceuticals after use is excreted in urine and faeces, and thus is present in municipal wastewater. However, wastewater treatment plants (WWTPs) in general have not been designed to deal with these compounds, as a result of which only 60-70% of the total load of pharmaceuticals and their metabolites is removed from WWTP effluent. The rest ends up in surface water, which in turn is used to produce drinking water. Research in the Netherlands has shown that WWTPs significantly contribute to the pharmaceutical concentrations in small surface waters[5], which leads to the conclusion that the presence of pharmaceuticals in surface water may become a problem for drinking water production. At the moment more than 4000 chemical compounds are being applied as pharmaceuticals, and in a small country like The Netherlands (with approximately 17 million people) yearly 3.5 million kg of pharmaceuticals are used, 140,000 kg of which end up in surface water. It is expected that these amounts will increase in the coming years, as a result of the development of new pharmaceuticals and of aging of the people [6]. Besides, due to climate change longer periods of draught are expected to occur, resulting in lower river discharge and thus higher concentrations. It already has been shown that some of these compounds, like diclofenac and hormone disruptors, also have a negative effect on the aquatic environment. Recently, the EU has put some of these compounds (17-Beta-estradiol (E2), 17-Alpha-ethinylestradiol (EE2), and

diclofenac) on a watch list, and it is expected that eventually standards will be set for these compounds [7, 8].

A separate category of pharmaceuticals are the veterinary pharmaceuticals. As these often are excreted with manure, they may enter the environment more diffusely, and may finally end up in groundwater [9]. This, however, may take several years, as a result of which it has to be kept in mind that, even after a certain type of pharmaceutical may have been banned still it may be observed in groundwater for many years.

Apart from the above-mentioned pesticides and pharmaceuticals also industrial compounds, originating from industrial wastewater treatment plants, can be found in surface waters. Their relevance for drinking water production depends on their concentrations and characteristics, which also determine their behaviour in water treatment processes. Unfortunately, very often little is known about their presence in wastewater or surface water.

Another category of pollutants are (micro)plastics. As in many cases this is particulate matter, other techniques will be required to deal with these materials. They cannot be degraded or adsorbed by common processes, and often the particles are too small (micro and nano scale) for removal in regular filtration processes like sand filtration. No standards have been set yet for water.

A recent trend observed, initiated by REACH registration, is that apolar chemicals are replaced by more polar ones [10]. More polar chemicals are better soluble in water and therefore removal in a treatment process is often more difficult.

1.4 Water quality: presence and characterization of Natural Organic Matter

Natural Organic Matter (NOM) is present in all surface ground and soil waters. It affects biogeochemical processes (like metal complexation and redox conditions) as well as water treatment processes through several mechanisms. Therefore, it is a key parameter with respect to design and operation of water treatment processes. It is responsible for color, taste and odor problems, and the major part of the coagulant and disinfectant requirements, it hinders the removal of other contaminants (e.g. by competition for adsorption sites in activated carbon, by pore blocking in filters, and by interference with photochemical and oxidation processes), it acts as a precursor for unwanted (disinfection) byproducts during treatment with chlorine and ozone, it contributes to membrane fouling, corrosion and the formation of metal complexes, and it acts as a substrate for bacterial growth, resulting in biologically unstable water and metal complexes [11-14].

NOM is a complex heterogeneous mixture of various organic molecules originating from the natural biological activity in water. Its composition varies from largely aliphatic to highly colored and aromatic, from highly charged to uncharged, with a wide variety of chemical compositions and molecular weights, depending on its origin [11, 15-17]. NOM originating from plant matter has a high lignin content, with a predominant aromatic fraction [18]. It is suggested that aromatic parts of wood and non-woody plants are the precursors of soil humic acids [19]. These authors describe that aromatic hydroxyl carboxylic acids and aldehydes are formed during UV and sunlight irradiation of lake and river NOM. Often NOM represents a family of polymeric chains resulting from the condensation of polyphenols [20]. Color often is caused by the presence of fulvic acids, humic acids and hyalomelanic acids.

The amount and character of NOM in water differ with climate and the hydrological regime as well as with other environmental factors. NOM found in natural waters consists of both hydrophobic and hydrophilic components. Approximately 50% of the Total Organic Carbon (TOC) in water consists of hydrophobic acids: humic acids, fulvic acids and humin. This fraction contains much aromatic carbon,

phenolic structures and conjugated double bonds. Hydrophilic NOM contains more aliphatic carbon and nitrogenous compounds like carbohydrates, sugars and amino acids. Dissolved Organic Carbon (DOC) content and TOC content often are used as indicators for NOM, but they give no information on its composition. The composition of NOM can be characterized in various ways, for example by dividing it in a soluble ($< 0.45 \mu\text{m}$, also containing cell fragments and macromolecules) and a particulate (suspended) fraction ($> 0.45 \mu\text{m}$). Soluble Microbiological Products (SMPs) end up in the water during e.g. biological treatment in municipal wastewater treatment plants. They originate either from the conversion of organic compounds by micro-organisms, or from dead micro-organisms. The SMPs contain humic acids, polysaccharides, proteins, amino acids, antibiotics, extra cellular enzymes, parts of micro-organisms themselves, and conversion products [21, 22].

A common method to characterize organic material is by means of LC-OCD (Liquid Chromatography-organic carbon detection) techniques [23]. In this case the following classification is applied:

- Biopolymers (BP) with molecular weight (MW) $>> 20.000$
- Humic substances (HS) with $\text{MW} \approx 1000$
- “Building blocks” (BB) with $\text{MW} \approx 300\text{-}500$ (These are natural conversion products of humic substances)
- Neutral components with $\text{MW} < 350$
- Acidic components (Low MW-acids) with $\text{MW} < 350$

Size exclusion Chromatography (SEC) often also is applied to determine the molecular weight distribution of the material.

Assimilable organic carbon (AOC) is a mixture of various fractions of organic material, which differ per type of water [24]. Grefte concluded that per type of water a specific linear relation can be observed between the average AOC concentration and the concentration of LMW acids.

Important parameters in the characterization of NOM are the aromaticity and the hydrophobicity of the material. Both variables are related. In literature, the material often is characterized by its Specific UV-absorbance (SUVA). Material with a high SUVA value in general contains much high molecular weight compounds, whereas low SUVA value material contains hydrophilic, low molar mass and low charge density compounds. Moreover, compounds with a high SUVA have a higher aromaticity and more unsaturated carbon bonds. However, size distribution and SUVA value are not necessarily related: it is possible that e.g. coagulation largely affects the SUVA value, but at the same time hardly affects the molecular weight of the DOC. Aedenaert et al. [25] used the UV absorption at a wavelength of 310 nm to determine the presence and amount of NOM.

Fluorescence (fluorescence excitation emission matrix; FEEM) too is applied to characterize dissolved organic material from a biological treatment process [26]. These authors studied a method to determine the polarity of various NOM fractions by means of the “polarity rapid assessment method” (PRAM). In this method water is extracted by means of various adsorbents (solid phase extraction, SPE). Apart from the hydrophobic surface of the material (and its aromatic character), also the molecular weight and molecular weight distribution play an important role. Column materials used for this technique are e.g. C_2 , C_8 and C_{18} , which show an increasing capacity for hydrophobic components. The most important parameter to characterize the various fractions is the difference in hydrophobic surface of the various components. Furthermore, dipole interactions and hydrogen bridging are used for characterization, for example by applying anion exchangers with NH_2 (a weak anion exchanger) and SAX (a strong anion exchanger). This method is affected by the pH and ionic strength [26-30].

The PRAM method differs from the commonly applied extraction using a XAD resin, as in the XAD method a low pH is applied and separation of the fractions is carried out in series instead of in a parallel execution. In XAD different fractions can be isolated and analyzed. Thus it is possible to determine a mass balance based on the XAD method, whereas this cannot be done using the PRAM results [26, 29].

1.5 Water quality: conditioning

Carbon dioxide and carbonate, and as a result pH, play a very important role in the characteristics of (drinking) water and water treatment.

Carbon dioxide dissolves in water according to equation 1:



Subsequently, carbonic acid dissociates into bicarbonate and carbonate, according to equations 2 and 3.



The dissociation constant for reaction 2 (K_1) is $4.5 \cdot 10^{-7}$ mol/L, and the dissociation constant for reaction 3 (K_2) is $4.7 \cdot 10^{-11}$ mol/L at 25 °C. From this it follows that at equilibrium:

$$pK_1 = \text{pH} + \log \left(\frac{[\text{H}_2\text{CO}_3]}{[\text{HCO}_3^-]} \right) = 6.35 \quad \text{eq. 4}$$

$$pK_2 = \text{pH} + \log \left(\frac{[\text{HCO}_3^-]}{[\text{CO}_3^{2-}]} \right) = 10.33 \quad \text{eq. 5}$$

Ca^{2+} may react with CO_3^{2-} , forming CaCO_3 , which has a very limited solubility:

$$K_s = [\text{Ca}^{2+}] \cdot [\text{CO}_3^{2-}] = 3.8 \cdot 10^{-9} \text{ at } 25^\circ \text{C} \quad \text{eq. 6}$$

The degree of super or subsaturation of calcium carbonate is expressed as the saturation index SI (also Langelier Saturation Index (LSI)), and is defined as (eq. 7):

$$SI = \log \left(\frac{[\text{Ca}^{2+}] \cdot [\text{CO}_3^{2-}]}{K_s} \right) = \text{pH} - \text{pH}_s \quad \text{eq. 7}$$

In equation 7 $[\text{Ca}^{2+}]$ is the Ca^{2+} concentration, $[\text{CO}_3^{2-}]$ is the carbonate concentration, and K_s is the solubility constant for CaCO_3 . pH_s is the equilibrium pH of water containing identical concentrations of Ca^{2+} and HCO_3^- .

$$\text{pH}_s = \text{pK}_2 - \text{pK}_s - \log([\text{Ca}^{2+}][\text{CO}_3^{2-}]) \quad \text{eq. 8}$$

In general, in The Netherlands it is strongly recommended that the pH of drinking water should be above 7.4 in order to prevent the dissolution of zinc from brass in taps, and of copper and iron from pipe materials. In this way also the dissolution of lead will be decreased in places where still lead piping is applied. Furthermore, the SI should be > -0.2 in order to prevent the dissolution of calcium carbonate from cement. This recommendation sometimes contradicts the pH requirements mentioned above [31]. For conditioning marble filtration may be applied. As many organic micropollutants are salts, the pH controls their degree of dissociation, and thus not only their solubility in water, but also their behavior in adsorption, photolysis and oxidation processes.

A way to describe the precipitation of calcium carbonate is by applying the Total Hardness (TH), the Theoretical Calcium Carbonate Precipitation Potential at 90°C (TCCPP₉₀) and the Practical Calcium Carbonate Precipitation Potential (PCCPP). The TH is defined as the total concentration of calcium and magnesium ions in water (in mmol/L, or in German Degrees (°D), 1 mmol/L corresponding to 5.6 °D). In the Netherlands it is recommended to apply a TH < 1.8 mmol/L, in order to increase customer comfort and decrease the environmental impact (use of detergents and energy requirements). However, in the Netherlands a minimum calcium concentration of 1 mmol/L is required, based on the fact that sufficient calcium uptake is essential for human health [32]. However, there is no evidence that the presence of calcium in drinking water would positively contribute to this [33-36].

Furthermore, it is recommended that TCCPP < 0.6 mmol/L and PCCPP < 0.4 mmol/L, in order to prevent precipitation of calcium carbonate, which results in higher maintenance costs and lower lifespan for apparatus like laundry machines and dish washers.

Under these conditions most organic micropollutants are soluble, as a result of which they are relatively difficult to remove from the water. Another point of attention are heavy metals. By applying milk of lime for softening or marble filtration for conditioning small concentrations of heavy metals may be introduced into drinking water. Precipitation of CaCO₃ doesn't contribute to the removal of e.g. organic micropollutants.

2 Water treatment: drinking water from groundwater sources

In many countries, groundwater is used as a source for drinking water production. Groundwater is in general relatively clean, although due to biogeochemical processes a wide range of compounds can be dissolved. The compounds present in the groundwater depend largely on the soil composition of the aquifer. Also, the well or borehole conditions can have influence on the water quality of the abstracted water. In specific circumstances, groundwater can be under influence of surface water. This can be the case in karstic aquifers where cracks in rocks result in a direct shortcut between the surface water and the groundwater. Sometimes, groundwater is deliberately put under influence of surface waters by application of river bed filtration.

In principle, groundwater can be aerobic, slightly anaerobic and deeply anaerobic. Aerobic groundwater is open to the atmosphere and thus contains oxygen. Most dissolved compounds will be present in an oxidized state. Under these conditions iron from the soil doesn't dissolve in the water. In order to produce drinking water, even aerobic groundwater is aerated to increase the oxygen concentration and simultaneously decrease the CO₂ concentration. The most important parameters that will have to be adjusted in this case are pH, calcium content, bicarbonate concentration and saturation index (SI). This is called "conditioning".

When the groundwater originates from sandy soil it often is lime aggressive, as a result of which cement pipe material may be dissolved (SI < -0.2) or enhanced corrosion of drinking water mains may occur. Due to several degradation processes carbon dioxide is present in the water, and as there is hardly any calcium present, the CO₂ concentration may be (much) higher than its equilibrium concentration. This is reflected in a large negative value of the SI. Air stripping will remove CO₂, but the pH and HCO₃⁻ concentration still will have to be adjusted. For example marble filtration may be applied for this purpose, dissolving the marble pellets and adding some hardness to bring the water near equilibrium.

Sometimes the groundwater contains a relatively high calcium concentration (in calcium rich aquifers). Hard waters are no threat for public health. Nevertheless, hardness can be a nuisance as warm water devices have a shorter life and require more maintenance due to scaling at higher

temperatures (Calcium carbonate solubility decreases with increasing temperatures), and for instance higher concentrations of detergents will be required. For these reasons water in many cases is softened.

As mentioned before, there are three types of groundwater: aerobic, slightly anaerobic and deeply anaerobic [37]. The treatment requirements for the three types differ considerably. Fig. 2 shows block schemes for typical groundwater treatment schemes.

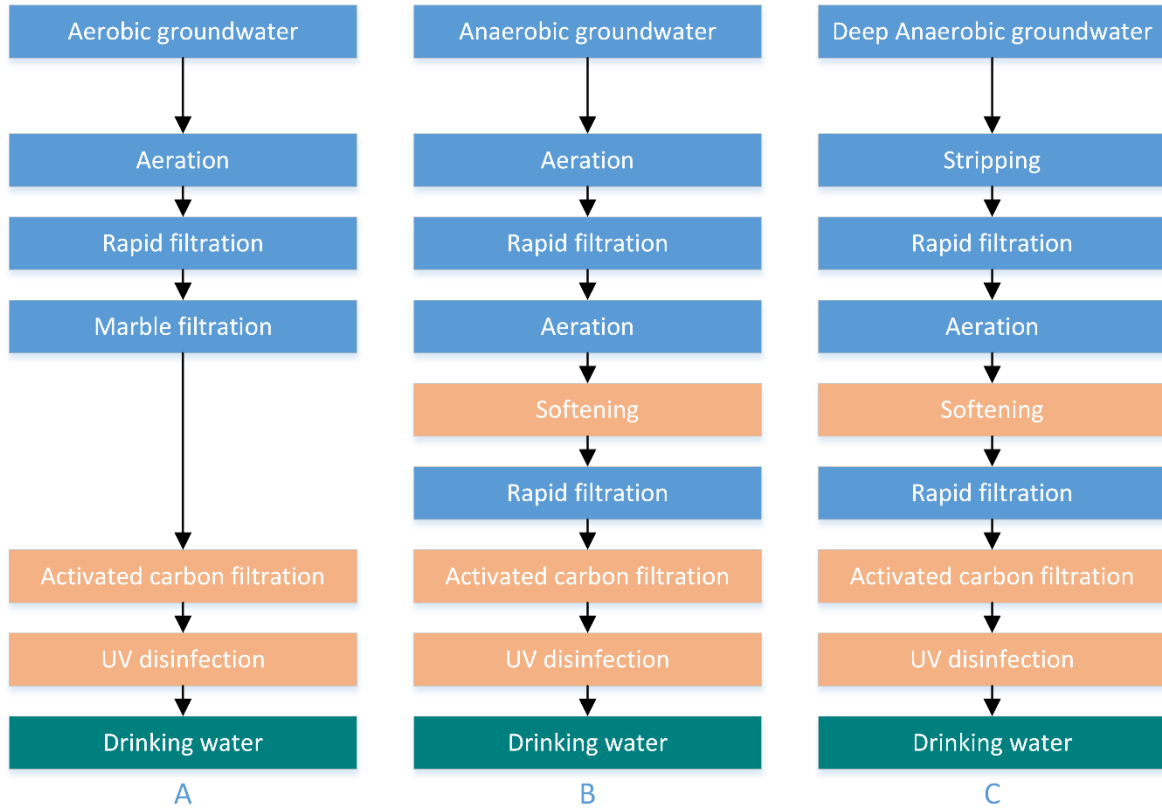


Fig. 2 Typical groundwater treatment schemes: A – Aerobic groundwater, B – Anaerobic groundwater, C – Deeply anaerobic groundwater. Orange blocks are optional, depending on the water quality: softening for hard water, activated carbon filtration and UV for the presence of organic micropollutants and disinfection.

The aerobic groundwater doesn't contain dissolved iron, but the anaerobic groundwater does. Besides, it contains ammonium and manganese. For slightly anaerobic groundwater aeration and stripping result in the removal of CO_2 , but also in the oxidation of Fe^{2+} to Fe^{3+} , of NH_4^+ to NO_3^- by biological processes, and of Mn^{2+} to MnO_2 , partly by chemical and partly by biological processes [38]. Fe^{3+} reacts with hydroxyl ions, forming $\text{Fe}(\text{OH})_3$ flocs, which slowly converts into iron oxide (Fe_2O_3). Bacteria, *Nitrosomonas* and *Nitrobacter* are responsible for the biological conversion of ammonium, which requires a relatively high amount of oxygen.

Deeply anaerobic groundwater may contain high concentrations of iron, manganese, ammonium, hydrogen sulfide and methane. Sometimes also chlorinated compounds, originating from industrial contaminations and spills, are found. Aeration and stripping are applied to remove the gasses and to oxidize iron, manganese and ammonium. For the nitrate formation "dry filtration" has to be carried out, in order to provide sufficient oxygen for the oxidation process. The removal of gasses can be described by applying Henry's law (eq. 9):

$$C_w = K_H * C_g \quad \text{eq. 9}$$

In which C_w is the equilibrium concentration of a gas in water, C_g is the equilibrium concentration of a gas in air, and K_H is Henry's constant or distribution coefficient. Stripping may also be an effective method to remove volatile contaminants, like vinyl chloride and 1,2-dichloro ethane, which sometimes are encountered in groundwater due to industrial pollution. However, most organic micropollutants are soluble salts, which cannot be removed in this way.

Simultaneously, iron and manganese are oxidized, forming precipitates. Colloidal particles, with a size $< 1 \mu\text{m}$, are kept floating by the Brownian motion. In case of neutral particles, stirring will result in collisions of the particles, upon which London –van der Waals forces will keep them together, forming larger flocs. These flocs may also include other species, like heavy metals (arsenic) and NOM/DOC. They precipitate, the rate depending on their size and composition. Rapid sand filtration then can be applied to remove the flocs from the water. Thus, also color, which originates from DOC, may be removed from the water.

Organic micropollutants, like pesticides and pharmaceuticals, resemble NOM in certain ways, and thus also may be included into the flocs. However, often these compounds are more hydrophilic and polar, as a result of which flocculation is not a very efficient process for the removal of these compounds. During the last decades more and more filtration over activated carbon (AC) has been applied to remove these compounds. AC is very effective for the removal of organic, and preferably hydrophobic compounds. As a result, serious competition is observed by NOM in the removal of micropollutants. Other problems that are encountered are pore blocking by NOM, as a result of which micropollutants cannot reach the pores anymore, and the effective surface for adsorption is reduced, and displacement of already adsorbed compounds by less soluble and more hydrophobic NOM compounds.

3 Water treatment: drinking water from surface water sources

Surface water has a much more dynamic behaviour than groundwater. Water quality can vary quite rapidly and river discharge flows can vary, in bigger rivers sometimes up to 3 orders of magnitude. Moreover, surface water contains a much wider variety of pollutants, as it is under direct anthropogenic influence by discharging of municipal and industrial wastewaters, surface run-off, atmospheric deposition, and incidental spills and calamities. It is therefore also impossible to control water quality. This puts additional pressure on a treatment system to produce safe drinking water.

Surface water treatment exists in a wide variety of systems and in recent years innovation in this area is accelerating. Nevertheless, a number of general treatment objectives have to be met: removal of suspended solids, disinfection, removal of taste and odour compounds, removal of a wide variety of micropollutants. Fig. 3 shows a number of more traditional surface water treatment schemes.

Suspended solids are mostly removed by application of coagulation, flocculation and sedimentation. Often these steps are followed by a rapid filtration. Disinfection traditionally was done chemically by adding chlorine. However, because of the detrimental health effects of the by-products, these systems are mostly abandoned nowadays. Other disinfections systems and multi-barrier approaches have been introduced. Many surface water treatment works also contain improved barriers against micropollutants often in combined processes for oxidation and disinfection like ozone and activated carbon filtration. Below the removal of suspended solids, disinfection and activated carbon filtration will be discussed.

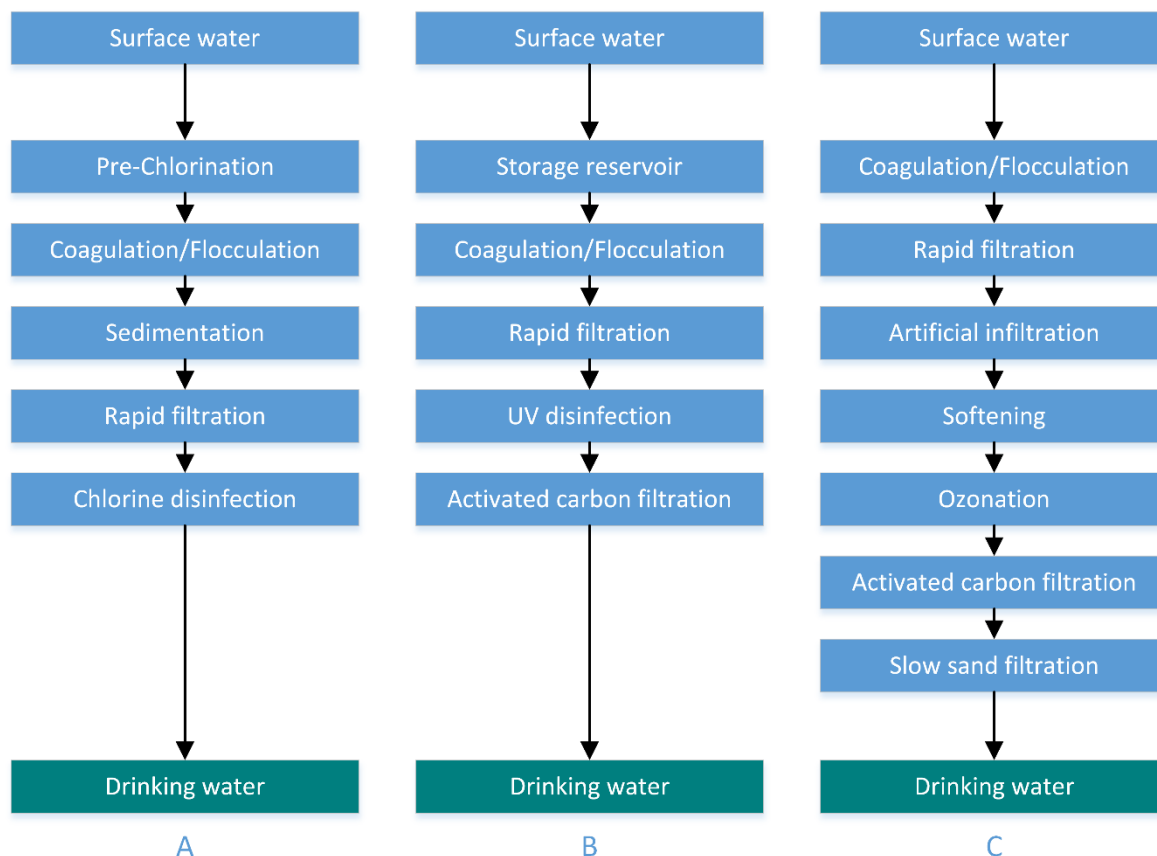


Fig. 3 Typical surface water treatment schemes. A: traditional, B: advanced, C: advanced including soil passage.

3.1 Coagulation, floc formation and floc removal

An important step in producing clean water is the removal of suspended matter. Suspended matter can consist of a large variety of materials and a wide range of particle sizes, e.g. clay and silt, organic debris, plastics and engineered nanoparticles. Part of the suspended matter may be stable in water, whilst other fractions may settle or float. This depends on the water chemistry, the density of the particulate matter relative to water, the temperature and viscosity of the water, flow and mixing etc. These conditions often vary hourly, daily and over the seasons.

Suspended matter causes turbidity in water, which is not only an aesthetic problem but also leads to poor treatment and disinfection performance. When the suspended matter content is relatively low the particles can often be removed by filtration. However, if the concentration increases, or if the particles are too small, a filtration step is in many cases insufficient or hindered by operational problems such as filter blocking. In that case, it is necessary to utilise other particle removal techniques like coagulation, flocculation and clarification. The suspended solids concentration that determines whether filtration or coagulation should be applied depends largely on the characteristics of the particulate matter and the water composition and is therefore difficult to predict.

Although coagulation, flocculation and clarification are different processes from a physico-chemical perspective, in practice they always are connected and taking place in the same unit operation. Coagulation is a process of destabilising the suspended particles by reducing the repulsive forces between them. This can be done by adding a salt or metal ion solution. The effect is that the particles start to form agglomerates. The agglomerate now starts to grow further into larger flocs. This process, called flocculation, needs gentle stirring to create collisions between the particles. Often a

flocculant or flocculation aid, such as a polymer solution, is added to increase the stability of the flocs. Once the flocs are large enough the suspension can be clarified by sedimentation or floatation.

Colloid stability

Suspended particles in water can span a large size range. The smallest particles are often a few nanometres and can have a natural origin (minerals, clays) or can be manmade (e.g. TiO_2 , n-Ag, ZnO, n- C_{60}), whilst the larger particles can be a few hundred micrometres. The latter can be of organic origin or larger sand grains. The smaller size particles or colloids are often quite stable in water, which means that the particles remain in suspension if the water is stagnant. They have a typical diameter of a few micrometres or less. Only larger particles, or the heavier ones will sink in a stagnant water body. Particles with a lower density than water, e.g. organic materials, oils, fats, will float on the surface.

Colloids in water can be stabilised by two independent physical principles. The first one is steric stabilisation (Fig. 4). Steric stabilisation occurs if the water contains water soluble polymers or large molecules like humic substances. These materials can adsorb on the particle surfaces. The length of the polymers prevents that the particles can approach each other at a sufficient short distance to form an agglomerate. However, depending on the polymer chain length and/or particle concentrations, the materials can start to form a stable network in the water.

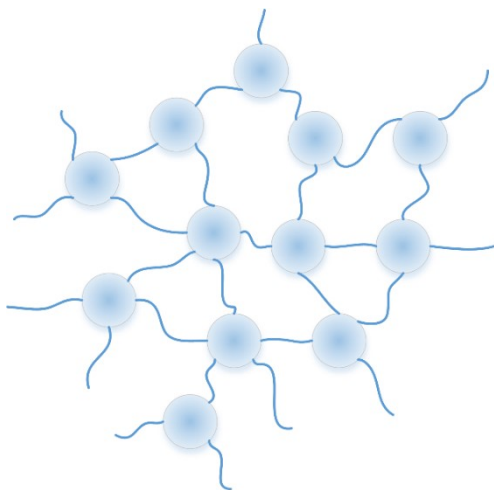


Fig. 4: Steric stabilisation

The second form of stabilisation can occur if the particle surface is charged. If a surface charge is present at the particle surface, an electrical double layer will occur around the particle (Fig. 5). This is caused by adsorption of ions from the water phase or ionisation of molecules at the particle surface. Ions of an opposite charge are attracted by this charge, thus forming an electric double layer [39]. In most cases particles in water, partly being clay minerals, are negatively charged. Only at very acidic pH positive surface charges may occur.

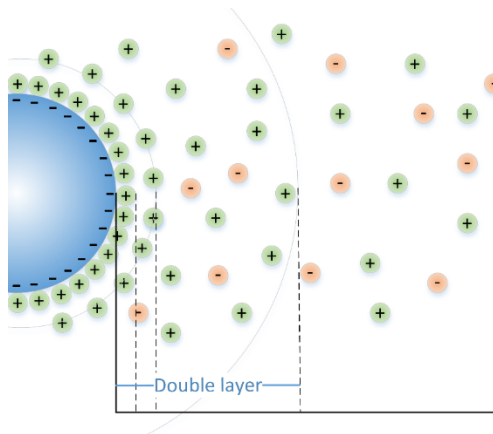


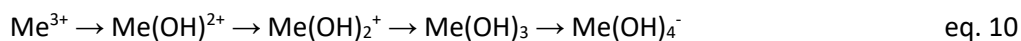
Fig. 5 Electric double layer

Also significant parts of the NOM, the humic acids, are negatively charged. Upon mixing the colloidal particles and compounds will repel each other, as a result of which coagulation will not occur. In this case the electrolyte content of the water can be increased. In this way, the double layer surrounding the colloidal particles may be compressed, decreasing the zeta potential of the surface [40]. It is also possible to include positively charged particles in the flocs by adsorption of highly charged counter ions, which also decrease the zeta potential. Thus, the repelling Coulomb forces can be decreased until they become smaller than the London-van der Waals forces.

Destabilization and floc formation

Suspended colloids can be destabilized by reducing the repulsive forces between the particles. Adding positively charged ions will reduce the electrostatic repulsive forces between the particles, because the positive ions will interact with negatively charged particle surface. Divalent ions appear to be about eight times as effective as a monovalent ion in causing coagulation, whereas a trivalent ion is even 600 times as effective [41]. For this purpose, in practice often Fe(III) or Al(III) salts are added to the water.

All metals cations are hydrated to some extent in water. In the primary hydration shell water molecules are in direct contact with the metal ion. The bonding of the water in the secondary hydration shell is more loose. For Al^{3+} and Fe^{3+} ions the primary hydration shell consists of six water molecules in an octahedral co-ordination. Due to the high charge on the metal ion the water molecules in this shell are polarized. This may lead to a loss of one or more protons, depending on the solution pH. As a result, the water molecules in the hydration shell are progressively replaced by OH ions, lowering the positive charge, according to equation 10:



In equation 10 Me is the metal (iron or aluminum). However, equation 10 is a simplified scheme, as in practice various polynuclear forms are observed [42]. Examples are $\text{Al}_2(\text{OH})_2^{4+}$ and $\text{Al}_3(\text{OH})_4^{5+}$, but equivalent species can be found for iron. Furthermore, also polynuclear hydrolysis products exist, like $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$. This is known as poly aluminum (PACl), and its chloride salt is a very effective flocculant.

At about neutral pH both Al(III) and Fe(III) salts show a limited solubility. As a result, amorphous hydroxide precipitates, which plays a very important role in the coagulation and flocculation processes. Positively charged precipitate particles may deposit on colloidal particles (heterocoagulation), resulting in charge neutralisation and thus destabilization [42]. In order to obtain effective coagulation a very short mixing time is beneficial, which means often high mixing

intensity is required. This can be realized by e.g. dosing in a cascade (Fig. 6) or utilizing a high shear turbine mixer. In the Netherlands mainly Fe-salts are applied, whereas in other countries Al-salts are preferred, sometimes combined with active silica [43]. An aspect which requires some attention is the pH of the salt solutions. Often they have a pH value ≤ 2 . By administering the solutions at one injection point, pH gradients may occur, with locally very low pH values. At these values certain hydrates can be formed, which are relatively stable, are ineffective adsorbents, and cause turbidity in the water. A high mixing rate is required to prevent the formation of such hydrates, and addition should take place at different locations. In practice the process starts at a relatively high mixing rate, which later is reduced in order not to destroy the already formed flocs by too high shear rates (tapered coagulation)



Fig. 6 Dosing of ferric chloride in a cascade

Another way to form larger particles is by adsorption and bridging of macromolecules and polyelectrolytes. This process is known as “flocculation”. For this purpose both natural and synthetic flocculants (mainly polyelectrolytes) can be applied. Anionic, nonionic and cationic polymer can be used. When the concentration of flocculant is too low, bridging cannot occur, and the floc size remains small. At too high flocculant concentrations the flocculant will cover the total particle surface, as a result of which bridging also cannot occur. In practice the addition of Fe^{3+} or Al^{3+} is often combined with the addition of a polyelectrolyte, thus enhancing flocculation. The use of polymers results in the formation of larger and stronger flocs with an open structure. The higher the molecular weight of the polymer, the more effective the flocculant will be. However, in this case too care should be taken not to apply a too high polymer concentration, as this would shield the colloidal particles or may cause so much steric hindrance that bridging of particles would be hindered. Sometimes addition of a salt will increase the adsorption of polymers by decreasing the electrical double layer. Addition of more particles, like clay particles, may also enhance flocculation, as it facilitates collisions between particles. Examples of flocculation tanks are shown in Fig. 7.



Fig. 7 Top: tapered flocculation tanks; Center: mixing device in flocculation tank; Bottom: flocculation tank in Wuppertal (Germany)

When this process occurs at a high rate other compounds or contaminants may be included in the flocs, which is called “sweep coagulation”. This process is more effective than for contaminant removal than only applying charge destabilization. Characteristics of the sweep coagulation process

are that flocs form more rapidly and can become much larger. Fig. 8 shows the working areas for pH and metal ion dose for the different modes of coagulation, such as sweep coagulation, adsorption destabilization and charge neutralization.

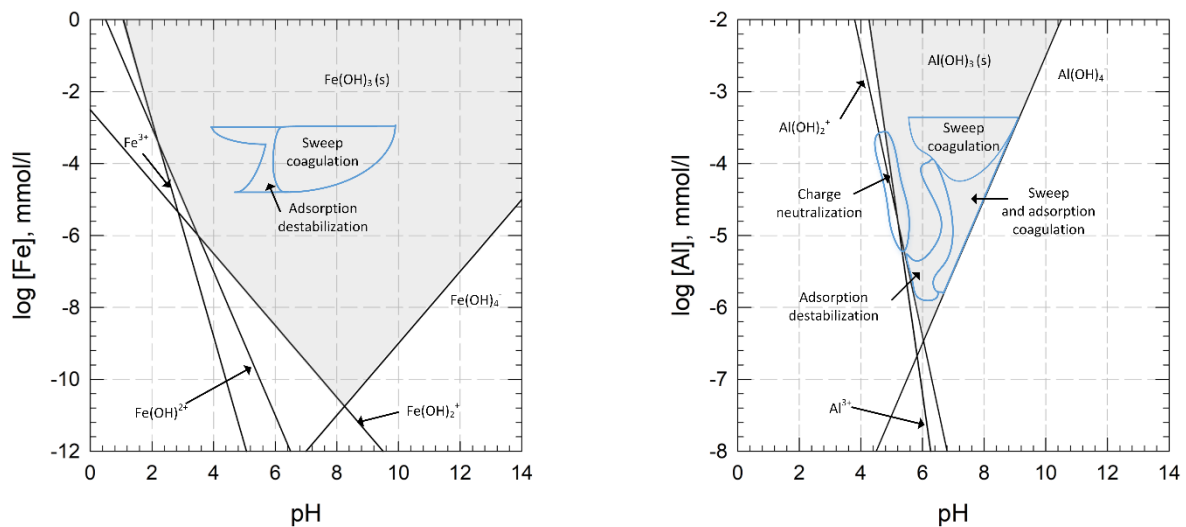


Fig. 8 Coagulation diagrams Alum and Ferric (adapted from [44] and equilibrium constants from [45])

When excess coagulant is used, more than would be required for baseline coagulation, this is called enhanced coagulation. The effect can be increased by changing the pH, the order of chemical addition or by using alternative coagulant chemicals. In this way TOC and NOM removal may be improved [11].

The time required to transport colloidal particles and flocculants is in the order of 10^{-4} s, whereas very high molecular weight polymers would require a few seconds. As a result good mixing is required to obtain optimum flocculation. pH may be an important parameter in this respect. Often it is found that within a certain pH range, in general $\text{pH} \approx 8$, a minimum flocculant dosage is required. In case this is a narrow range it is difficult to maintain the right pH during the process. This also depends on the color of the water: in general it is found that the more color there is present the lower the optimum pH will be. In order to remove fulvic acids a higher dose of flocculant is required than for the removal of humic acids, which probably is caused by the lower molecular mass of the fulvic acids.

During coagulation and flocculation iron(hydr)oxides are formed, which precipitate (see Fig. 9). It is known that these compounds can react with phosphate. As a result, during flocculation also phosphate is removed from the aqueous phase. The same principle is encountered for arsenic, which also can (partly) be removed by flocculation in this way. However, for other heavy metals the method is less effective.



Fig. 9 Horizontal sedimentation

Pollutant removal

Organic micropollutants may be removed by flocculation, but only if they have been adsorbed to some other, high molecular weight compounds. This will, amongst others, depend on pH, which determines the charge of the molecules, and thus their ability to be adsorbed. In general, removal of organic micropollutants by flocculation is very limited, as was demonstrated by van der Horst et al. [46] for pharmaceuticals and endocrine disrupting compounds (EDC's) and by Saraiva Soares et al. [47] for pesticides and their metabolites (endosulfan, ethylenethiourea (ETU), and 1,2,4-triazole). This mainly is due to the solubility and hydrophilicity of the micropollutants.

Micro-organisms like bacteria, and algae may be removed by sweep coagulation and by bridging. For viruses removal may occur by complexation with aluminum and iron. However, the level of removal in general is insufficient to obtain safe drinking water [48].

Recently, in a case of an industrial contamination of river water, used for the production of drinking water in the Netherlands, it has been found that in a rapid sand filter (Fig. 10) micropollutants may be removed by means of biodegradation. However, the effectiveness of this biodegradation strongly depends on circumstances (like temperature), and the presence of certain types of micro-organisms.



Fig. 10 Rapid sand filtration in the Netherlands (left) and in Germany (right)

Floc removal

Once the flocs are formed, they have to be separated from the water stream. In most cases this is done by sedimentation or gravity settling. In practice, many different forms of sedimentation tanks exist. The simplest form is the long rectangular sedimentation pond as shown in Fig. 9. The length of

the tanks should be such that under the low flow conditions the residence time is sufficient for particles to settle to the bottom of the tank. Periodically the sludge is removed from the tanks by dredging. As these horizontal tanks need a large surface area, other engineering solutions such as lamellae separators and circular clarifiers have been designed. Instead of applying sedimentation to remove suspended matter also flotation can be applied, especially in case of low density particles or e.g. oil droplets or algae (Dissolved Air Flotation; DAF; see Fig. 11). In this case air is dissolved under pressure, and then by means of nozzles it is released in a tank at atmospheric pressure. As a result small bubbles (10 - 100 μm , on the average 40 μm) will be formed. In order to ensure the formation of such small bubbles the pressure differences should be 400-600 kPa [49]. These bubbles may adsorb surfactants and/or NOM and thus obtain a negative charge. Bubbles may adsorb hydrophobic particles. By dosing a coagulant like polyaluminium chloride or ferric chloride particles will be destabilized, promoting the adherence of the bubbles to the particles, causing them to float to the surface, where they can be removed by means of skimming. Care should be taken not to overdose the coagulant, as this will result in charge restabilization of the positively charged particles and bubbles. It is possible that organic micropollutants also will be removed in this way, but the method is not very effective, as these compounds often have a high solubility.

As a final step to remove residual suspended solids, but also to remove excess coagulants, a rapid filter step is applied. These filters also contribute to a multi-barrier disinfection.



Fig. 11 Dissolved air flotation in Norway

3.2 Application of activated carbon

Activated carbon often is used to improve the water quality by removing natural organic matter and organic micropollutants. Two types of activated carbon can be applied:

- Powdered activated carbon (PAC)
The particle size of this material is $< 0.05 \text{ mm}$. It is added to the water, where it can adsorb organic compounds, like NOM but also organic micropollutants. As it has a high surface area, due to its small particle diameter, filtration requires a short contact time of about 10-15 minutes. Afterwards the activated carbon has to be removed by means of filtration.
- Granular activated carbon (GAC)
The granules have a diameter of 0.3 – 3 mm, and have a porous character. The pores contribute to the adsorption capacity of the material. However, pore size (micropores $< 1 \text{ nm}$, mesopores 1 – 25 nm, and macro pores $> 25 \text{ nm}$) determines which compounds can enter

the pores. Another aspect of the pores is that they may be blocked by NOM which adsorbs at the surface. After the GAC has been loaded it can be reactivated, by heating under an inert atmosphere, followed by activation, e.g. by an acid. GAC is applied in filter beds (Fig. 12).

The adsorption properties of the activated carbon strongly depend on the physical characteristics of the carbon (pore volume and size, surface area), and the chemical characteristics (surface composition, which depends on the activation method applied). Furthermore, in GAC filters micro-organisms may develop, as a result of which biodegradation also will take place inside the column. This improves the removal of organic compounds by the filter, and results in a longer time to reactivation of the GAC.

Whether organic micropollutants can be efficiently removed by activated carbon strongly depends on the micropollutant characteristics. In general, it can be assumed that the more hydrophobic the compounds are, the higher the adsorption capacity of the carbon will be. However, NOM may act as a competitor. Other parameters that may affect the effectiveness of activated carbon are temperature, pH and the presence of salts. Small, hydrophilic compounds in general are very difficult to remove by means of activated carbon.

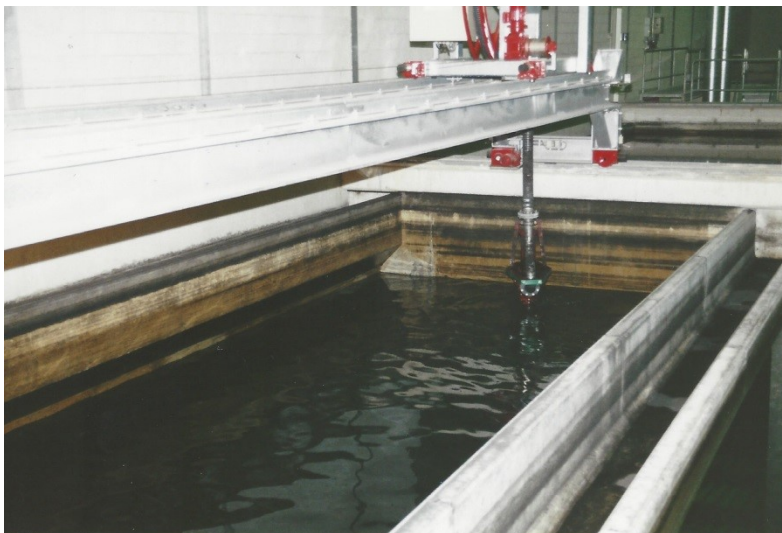


Fig. 12 GAC filter at Leidsin (The Netherlands)

After some time, the activated carbon will be saturated and adsorption will stop. For powdered activated carbon, an equilibrium between the adsorbed and dissolved compounds will occur quickly after dosing it. The PAC can therefore be used only once and has to be removed after dosing. For granular activated carbon, the adsorbing compounds will gradually saturate the filter column. During operation, a mass transfer zone will exist in the filter that slowly moves down in the flow direction in the filter bed. After some time, the compound will break through and the filter column has to be taken out of operation and the activated carbon has to be regenerated.

3.3 Disinfection

In the 19th century it was discovered that some compounds, like chlorine, can be used as a disinfectant. Since the beginning of the 20th century disinfectants were applied by drinking water companies. However, since the 1970s it became clear that by adding chlorine (toxic) disinfection byproducts may be formed by the reaction of chlorine with humic acids [50]. Because of this ozone became more popular as a disinfectant. However, also ozone appeared to give harmful byproducts, as it reacts with bromide to form bromate.

Membrane filtration also can be applied, but also appeared to have some disadvantages: fouling of the membrane surface may occur and sometimes leakage is observed, decreasing the disinfection effectiveness.

In Marseille in 1901 UV was applied for disinfection purposes, but it took about 50 years until it was applied on a larger scale in Switzerland, Austria, and Norway. Since the last quarter of the 20th century application of UV for water disinfection has become quite common.

The photochemical active part of the electromagnetic spectrum can be divided into four regions: vacuum UV (VUV; 100-200 nm), UV-C (200-280 nm), UV-B (280-315 nm) and UV-A (315-380 nm). This division is based on physical, biological or medical parameters.

UV-C radiation appears to be absorbed by DNA and RNA in cells of organisms. This may result in the death of these cells, or will prevent their reproduction. The term “inactivation” can be applied to either a cell or its single subsystem. UV irradiation has been shown to be a powerful tool in inactivation of both microorganisms and cells such as bacteria, viruses, protozoan parasites, some spores, living cells, and subsystems such as enzymes, aminoacids, and lipids [51]. One of the main advantages of UV inactivation is that no chemicals have to be added, since irradiation is a physical process. The germicide effect of UV light on bacteria and viruses is primarily due to the formation of thymine, thymine-cytosine (pyrimidine) and cytosine dimers in polynucleotide chains of DNA (they are listed in prevalence of order). It seems that radiation at wavelengths in the range of 200-295 nm (so called bactericide or germicide range) exerts the most effective action. The absorption spectrum of DNA of viruses and bacteria shows a maximum at about 260 nm. However, also at wavelengths between 115 and 160 nm the absorption coefficient of DNA is high [52].

For killing microorganisms it is not always necessary to change the DNA of the cells. It may also be enough to damage the cell membrane, allowing other compounds to enter the cell and damage or kill it. (V)UV photons with a wavelength below 275 nm can break C-C (3.8 eV) or C-H (4.5 eV) bonds, thus damaging the cell membrane or proteins in the cell. Furthermore, photo desorption producing species eroding the outer coat of the spore may lead to cell death during germination. Cells can be destroyed by etching [53]. Sosnin et al. also distinguish between two different disinfection methods: the inactivation of microorganisms by UV irradiation or their total VUV-induced photo mineralization

Bacterial spores, which are the most resistant form of living microorganisms, often are used as model microorganisms in studies [52]. The reason that spores are 10-50 times more resistant to 254 nm UV light than their corresponding growing cells, is the presence of a unique UV photoproduct of the spore DNA (called SP) [54]. Finally, spore DNA is protected by multiple layers, which surround the core, namely a germ cell wall, cortex, inner and outer spore coats, and sometimes an exosporium. In *B subtilis* spores these form a 150-200 nm thick proteinous barrier, which can shield the core from the effects of (V)UV photons. It is possible that UV radiation can also kill spores by modifying these outer layers of the spores, but the importance of this second pathway is not yet fully known.

Templeton, Andrews and Hofmann [55] have shown that humic acid flocs and particles < 2 µm may shield viruses from UV radiation, thus decreasing the disinfection effectiveness of a UV process.

In order to inactivate micro-organisms it has been established that it isn't necessary to kill them. As a result, UV doses applied for disinfection have been decreased in the past years, in order to decrease the energy demand of the process. At the moment in general a UV dose of 20-70 mJ/cm² is applied in order to obtain effective disinfection of drinking water. In principle, photolysis by UV irradiation can also degrade organic micropollutants. This depends on the wavelength, i.e. the energy of the UV photon that can be absorbed by the molecule. In practice two types of mercury containing UV lamps

are being used: low pressure (LP) UV lamps, which emit a single wavelength of 253.7 nm, and medium pressure (MP) UV lamps, which emit a much broader spectrum between 200-300 nm. Obviously, MP lamps will be more effective in causing photolysis of micropollutants because of this broader spectrum. However, it has been shown that for effective photolysis, much higher doses will be required than is common for disinfection purposes [56, 57]. Therefore, it can be concluded that UV disinfection in general will contribute very little to the degradation of organic micropollutants.

4 Water treatment: drinking water from river bank filtrate or dune filtrate

Sometimes river bank filtrate is used instead of groundwater. By filtration through soil the water quality can significantly be improved, partly by adsorption to soil particles, partly as a result of biodegradation by sub-soil micro-organisms. In this way also some micropollutants may be removed, depending on the local conditions (like the type of soil and water, presence of micro-organisms, molecular properties of the micropollutants, etc.). However, not all micropollutants appear to be removed in this way. Because of this the treatment process may be simpler than the treatment process required for surface water treatment. In general aeration, in order to increase the oxygen content, sand filtration, conditioning and disinfection will be applied, but, depending on the water quality, other techniques, like filtration over activated carbon, also may be necessary.

A type of treatment typical for the Netherlands is dune filtration. This resembles river bank filtration, but in general the residence time is longer. Dune filtration significantly contributes to the removal of pathogens, but also of other compounds. To make dune filtration possible the high water quality is already required to protect the dune ecosystem. Especially organic micropollutants have to be removed before infiltration. For the final treatment similar processes as in case of river bank filtrate will be necessary.

5 Conclusions

Water is treated to meet the objectives of drinking water quality and standards. These objectives are to prevent acute diseases by exposure to pathogens, to prevent long-term adverse health effects by exposure to chemicals and micropollutants, and finally to create a drinking water that is palatable and is conditioned in such a way that transport from the treatment works to the customer will not lead to quality deterioration.

Traditional treatment technologies are mainly designed to remove macro parameters such as suspended solids, natural organic matter, dissolved iron and manganese etc. Which kind of technologies are required depends on the drinking water source. For groundwater in general aeration, filtration, conditioning and disinfection will be required, whereas for surface water coagulation/flocculation, sedimentation, several filtration processes, conditioning and disinfection will be necessary. These technologies have however only limited performance for removal of micropollutants. Advancing analytical technologies and increased and changing use of compounds show strong evidence of new and emerging threats to drinking water quality. Therefore, more advanced treatment technologies are required, in order to guarantee the production of safe drinking water.

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